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Biomass gasification gas cleaning for downstream applications: A comparative critical review

Mohammad Asadullah *

Faculty of Chemical Engineering, Universiti Teknologi MARA, 40450 Shah Alam, Selangor, Malaysia

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ABSTRACT

Biomass is the only source on earth that can store solar energy in the chemical bond during its growth. This stored energy can be utilized by means of thermochemical conversion of biomass. Gasification is one of the promising thermochemical conversion technologies, which converts biomass to burnable gases, often termed as producer gas. Major components of this gas are hydrogen, carbon monoxide and methane. Depending on the purity, this gas can be used in the furnace for heat generation and in the internal combustion engine and fuel cell for power generation or it can be converted to liquid hydrocarbon fuels and chemicals via the Fischer–Tropsch synthesis method. Despite numerous applications of the biomass gasification gas, it is still under developing stage due to some severe technological challenges. Impurities such as tar, particulate matters and poisonous gases including ammonia, hydrochloric acid and sulfur gases, which are unavoidably produced during gasification, create severe problems in downstream applications. Therefore, the cleaning of producer gas is essential before being utilized. However, the conventional physical filtration is not a technically and environmentally viable process for gasification gas cleaning. The utilization of catalyst for hot gas cleaning is one of the most popular technologies for gas cleaning. The catalyst bed can reform tar molecules to gas on the one hand and destroy or adsorb poisonous gases and particulates on the other hand, so as to produce clean gas. However, numerous criteria need to be considered to select the suitable catalyst for commercial use. In this review, the advantages and disadvantages of different gas cleaning methods are critically discussed and concluded that the catalytic hot gas cleaning with highly efficient catalyst is the most viable options for large-scale production of clean producer gas.

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* Tel.: +60 3 5543 6359; fax: +60 3 5543 6300.
E-mail address: asadullah@salam.uitm.edu.my

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1. Introduction

Biomass is one of the most plentiful organic materials on the earth, which is produced by photosynthesis reaction in green plant in the presence of sunlight. It stores solar energy in its chemical bonds as a chemical energy [1], which can be further evolved by breaking down the bonds [2]. Thermochemical conversions including combustion, gasification and pyrolysis are the processes that can break down the chemical bonds in biomass to release the stored energy. Combustion can directly release the energy by primary bond breaking of biomass, while gasification and pyrolysis can transfer the energy into secondary products (gas and liquid), which are likely to be ideal for fueling the furnace and the engine [3]. Based on the advantages in terms of energy efficiency and ease of application, gasification is the best choice for exploiting energy from biomass [4]. It converts biomass into fuel gas (producer gas), consisting of hydrogen, carbon monoxide, carbon dioxide and nitrogen as major components along with some methane and other minor components. This gas is readily burnable either in the furnace for heat generation or in the internal combustion engine for power generation [5–7]. Since the gas is rich in H_2 and CO, they can be separated to utilize for fuel cell [8,9] or to convert into liquid hydrocarbon fuels or chemicals by the Fischer–Tropsch synthesis method [10,11].

Despite the numerous advantages of biomass gasification, the technology is still in the developing stage due to some challenges. Impurities such as tars, particulate matters, NH_3, H_2S, HCl and SO_2, which are unavoidably produced during gasification and generally sustained in the producer gas, cause severe problems in downstream applications [12–16]. These contaminants must be removed before the gas is being used for internal combustion engine, fuel-cell, and for secondary conversion into liquid fuels or chemicals by Fischer–Tropsch synthesis [8–11]. Among the impurities, tar is the notorious one, which represents a number of organic compounds, especially aromatic compounds heavier than benzene [17–19]. Tar is a sticky material, which usually condenses in the low-temperature zone of the downstream applications and blocks the narrow pipeline. As reported, the tar tolerance limit varies depending on various applications such as $\sim$500 mg/Nm^3, $\sim$100 mg/Nm^3 and 5 mg/Nm^3 and is recommended for compressors, internal combustion systems, and direct-fired industrial gas turbines, respectively [20]. For Fischer–Tropsch synthesis, the tar concentration must be even lower ($<$0.1 mg/Nm^3) [21,22] along with ammonia concentration $<$10 ppm, which is produced generally in the range of 1000–5000 ppm in producer gas, depending on the raw materials and operating conditions used [23]. During gasification, most of the nitrogen content in biomass ends up as NH_3, N_2, HCN and HNCO as well as NO_x [23–26].

The formation of tar and NH_3 is a function of air–fuel ratio as well as process temperature. It is well reported that the higher air to fuel ratio and temperature favor reducing the tar and NH_3 concentration in the producer gas [18,27–29]. However, two problems can be encountered for high-temperature and high air–fuel ratio. Firstly, high-temperature gasification requires expensive alloy materials for reactor constriction as well as high temperature is very tough to maintain [30,31]. Secondly, the high air to fuel ratio reduces the burnable gas composition in the producer gas [32]. This means that the contaminants must be removed by other means such as physical filtration, wet scrubbing or catalytic hot gas cleaning. The physical filtration is a simple method of tar and particles separation; however, the agglomeration of sticky tar and particles often blocks the pores of filter. In addition, it cannot separate gaseous impurities. The most severe problem of physical filtration and wet scrubbing methods is the handling and disposal of toxic tar. For large-scale gasification of biomass, the stringent environmental regulation does not allow the disposal of such a huge quantity of toxic tar into the environment. Therefore, catalytic hot-gas cleaning could be considered as an attractive option for removing contaminants from the gasification gas. This method is indeed more advantageous in terms of energy efficiency as it eliminates the gas cooling step for physical filtration and the reheating step of gas for downstream application.

Comprehensive researches have been conducted for catalyst development in order to reform tar to gases over the last couple of decades. Tar is a mixture of a wide range of aromatic hydrocarbons and their derivatives. In principle, these aromatic hydrocarbons along with light aliphatic hydrocarbons including methane can undergo reforming or cracking reaction on some catalysts to form gaseous products at certain temperatures [33–37]. At the same time ammonia can also be decomposed on the Fe, Ni and Ru based catalysts [38–41]. However, HCl, H_2S and SO_2 do not decompose on the catalyst; instead they are highly soluble in water, and hence they can be separated by water scrubbing [42].

The reactions involved in catalytic hot gas cleaning are extremely slow due to the inertness of the poly-aromatic compounds, which is usually formed by the recombination of small molecules [43], requiring high temperature and activation energy to start the reaction. In addition, other gaseous impurities especially HCl, H_2S and SO_x can be permanently adsorbed on the active sites of the catalyst, so as to reduce the catalytic activity. Under the reaction conditions, the tar can be readily converted to coke, which in addition to particulate matter builds up on the catalyst surface and covers the active sites, hindering the tar and reforming agents to come into contact with the reaction site. Therefore, it is obvious that catalytic hot gas cleanup requires a highly reactive and resistive catalyst. The catalyst must be highly selective to gas formation rout instead of coke formation rout. In addition, the catalyst must be able to transfer oxygen to the deposited carbon to clean up the surface by fast oxidation reaction.

Different types of catalysts have been proven to be active for tar and ammonia decomposition. In order to reduce the tar content in the product gas stream, catalysts have been used either in the primary bed or in the secondary bed. In the case of primary bed, the catalyst is placed in the gasification reactor where the biomass is directly fed [44–46]. However, the catalyst is rapidly deactivated due to the fouling of ash and carbon on the surface. Non-metallic catalysts such as dolomite and olivine show longer activity in the

primary bed application; however, they are eroded and elutriated from the bed [44]. It is reported that the noble metal catalysts such as rhodium (Rh) can almost completely convert tar and char at unusually low temperatures (500–700 °C) both in primary and in secondary bed reactors [47–54]. However, as shown in the scanning electron microscopic images of spent catalyst (Rh/CeO_2), it was sintered during reaction [47–49]. The sintering problem was overcome when CeO_2 and Rh were loaded on porous silica sequentially as $Rh/CeO_2/SiO_2$ [50–54]. However, these catalysts still need to be investigated for long-run experiments.

Nickel based and modified nickel based catalysts are widely investigated for tar cracking in the secondary bed reactor [55–57]. These catalysts show superior activity for tar destruction; however, the catalysts cannot sustain until a desired length of time. Char-supported iron catalysts have recently been developed, which have shown superior activity in tar reforming. The tar concentration reduced to below 100 mg/Nm^3, which is the requirement for internal combustion engine for power generation [58–64]. Compared to silica and alumina-supported noble metal and nickel catalysts, char-supported iron catalysts are obviously cheap. Most importantly, the spent catalyst can be gasified to recover the energy from char, while iron can be recovered from the ash for further application.

From the above study, it can be realized that the cleaning of producer gas is essential and the catalytic destruction of tar is the most convenient way, which is supposed to provide higher overall efficiency of the process. However, the selection of catalyst is a real challenge, because of the numerous criteria to be considered. This review highlighted the advantages and disadvantages of different gas cleaning methods including physical filtration, thermal hot gas cleaning and catalytic hot gas cleaning in order to meet the quality of producer gas to be used in different downstream applications.

2. Biomass gasification gas impurities

In the gasification of biomass, not only burnable gases but also some unwanted impurities form including tar, particulate matter, NH_3, HCl, NO_x, H_2S, and SO_x. As described in the previous section, these impurities are real challenges in utilizing the producer gas. The quantity of impurities generally produced in different gasification methods and their effects in downstream applications are discussed in the subsequent sections.

2.1. Level of impurities in the producer gas from different types of gasifiers

The concentration of impurities in the producer gas depends on many factors; however, reactor types and gasification conditions are two major factors that control the producer gas quality. Scheme 1 exhibits the composition of product gases and impurities produced from different types of gasifiers. It is reported that the maximum tar yield can go up to 6 g/Nm^3 for air blown fixed bed co-current reactor, while it is 10–150 g/Nm^3 for counter-current reactor [65–68]. In co-current reactor, the gasifying agent and feedstock bring into contact at the inlet zone where the volatiles start to react with reagents and continue while they travel to the end of the reactor. Therefore, the left-over tar could be lower. However, the counter-current reactors feature oppositely. The feedstock inlet of the gasifier is close to the gas outlet at the top and the gasifying agent is introduced at the bottom. The air contacts first with solid char at the bottom and continues reacting with char and being volatile while it travels upward. The oxygen concentration becomes lower while the tar concentration is higher at the top of the reactor, and thus a higher amount of tar exits the reactor. Meanwhile, the particulate matter content in the producer gas is lower in the case of counter-current reactor than that of the co-current one. This is because the dust is normally formed at the end of the particle's reaction, which is closed to the outlet for the co-current reactor. In addition, the gravitational force allows the particles to exit from the bottom for co-current reactor. On the other hand, the gas composition also differs from each other. Because the steam generated from the devolatilization of feedstock travels concurrently in the co-current reactor with organic volatiles and product gas, while it further takes part in water gas shift reaction and steam reforming reaction to produce more H_2 along with other gases, which contribute to higher H_2 and CO_2 composition in producer gas, compared to the counter-current reactor. The LHV of producer gas produced in the co-current reactor is higher (maximum average 5.6 MJ/Nm^3), because of the higher total burnable gas yield compared to the counter-current reactor (maximum average 5.1 MJ/Nm^3).

Compared to fixed bed gasifier, fluidized bed gasifier, especially circulating fluidized bed gasifier, needs a high speed of air. Because of the short residence time of tar molecules in the reactor, the unconverted tar is much higher in the case of circulating fluidized bed reactor than that of fluidized bed gasifier [67–70]. However, compared to the counter-current fixed bed reactor, the tar is lower in producer gas from both fluidized bed gasifiers [65–67]. This is because, in the fluidized bed gasifier, there is enough free board for tar to convert, compared to the counter-current gasifier [71]. The dust particles loading in the producer gas are normally very high for fluidized bed gasifier, especially for circulating fluidized bed gasifier because of the high entrainment of the particles [67,69,70].

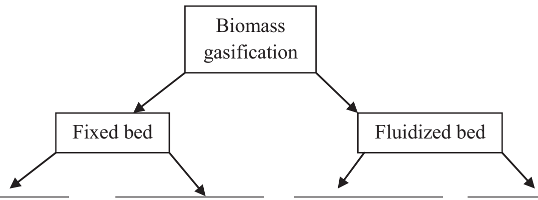
2.2. Effects of impurities in downstream applications and gas quality requirement

Different types of impurities including tar, particulate matter, NH_3, HCl, H_2S, and SO_2 in the producer gas affect differently in the downstream applications depending on their physical and chemical properties. The details of their effects and maximum acceptable level of their concentration in different downstream applications are summarized in Scheme 1 and critically described in the subsequent sections.

2.2.1. Particulate matters

Biomass feedstock inherently contains some minerals, which in gasification are converted into ash mostly in micron size amorphous form. In addition, some unconverted carbonaceous materials also form dust in micron size. Because of the fineness of the particles, it is very difficult to effectively separate from the product gas stream by conventional cyclone separator, usually integrating at the exit of the gasifier. Hence a significant amount of particles always exist in the producer gas. The extent of particle loading in the producer gas entirely depends on the gasifier design. The gas from the fluidized bed generally contains a higher loading of particles than that of fixed bed gasifier. When the producer gas is used for internal combustion engine, the particles deposit in the nozzle and other places and block the system. For turbine application the particles adversely affect the turbine blade due to abrasion effect. The particles also affect the anode of the solid oxide fuel cell and deactivate the catalyst for the Fischer–Tropsch synthesis. Moreover, the particles finally remain in the flue gas of IC engine and turbine and exceed the emission limit of environmental regulation.

Based on many studies, the particle loading limit in the producer gas is strictly imposed as shown in Scheme 1 and it is varied based on application. The internal combustion engine can satisfactorily accept the particle concentration <50 mg/Nm^3 with



Gas quality	Co-current [65-68]	Counter current [65-68]	Bubbling fluidized bed [88-92]	Circulating fluidized bed [67, 69, 70]
Tar, mg/Nm^3	10-6000	10000-150000	1500-9000	9000-10000
PM, mg/Nm^3	100-8000	100-3000	12000-16000	7000-12000
LHV, MJ/Nm^3	4.0-5.6	3.7-5.1	3.5-5.0	3.6-5.9
H_2, vol%	15-21	10-14	10-15	15-17
CO, vol%	10-22	15-20	13-20	15-18
CO_2, vol%	11-13	8-10	17-22	16-18
CH_4, vol%	1-5	2-3	1-4	4-6
C_nH_m, vol%	0.5-2	nd	nd	1.0-1.5
N_2, vol%	rest	rest	rest	rest

Gas quality requirement

Gas quality	IC engine [20, 72, 93, 94]	Gas turbine [20, 73, 95, 96]	F-T synthesis [97-99]
Tar, mg/Nm^3	< 100	< 5 (all in vapor phase)	< 1[a]
PM, mg/Nm^3	< 50	< 20	0
Particle size, μm	< 10	< 0.1	
Minimum LHV, MJ/Nm^3	-	4-6	-
Minimum H_2 content, vol%	-	10-20	-
Max alkali concentration, ppb	-	20-1000	< 10
S component (H_2S, SO_2, CS_2, ppm	-	< 1	< 1
N component (NH_3 + HCN), ppb	-	-	< 20
HCl, ppm	-	< 0.5	< 0.1
Alkali metals, ppb		< 50	< 10

[a]Unit in ppmV

Scheme 1. Producer gas quality produced from different gasifiers and the requirement of gas quality for different downstream applications.

size < 10 μm, while it is < 30 mg/Nm^3 for gas turbine [20,72–74]. The particles are essentially to be completely separated for Fischer–Tropsch synthesis for methanol or hydrocarbon fuel production.

2.2.2. Tar

Tar is a complex mixture of mostly aromatic hydrocarbons which are condensable at ambient temperature. This tar forms during the secondary reaction of devolatilized organic compounds with gasifying agents and generally exists in the producer gas stream. The tar can be further defined as a complex mixture of organic molecules with molecular weights greater than benzene [20,75,76]. From the definition, it can be realized that tar is condensable and it generally condenses at the low-temperature area in the downstream applications, resulting in plugging and fouling of pipes, tubes, and other equipment. The producer gas temperature for internal combustion engine is one of the crucial factors. The temperature is to be essentially below 100 °C for two basic reasons. First, to maintain the air–fuel ratio before feeding into the combustor, the fuel density must be as high as possible, meaning that the temperature must be as low as possible. Secondly, the moisture that is inherently produced in the gasifier essentially needs to be removed, which requires the temperature to be lowered down to below 100 °C. When the feed gas temperature is lowered down to ambient temperature, the tar begins to condense and become sticky materials, creating severe problems. Contrary to the fact that the moisture in the producer gas can be spontaneously condensed down at ambient temperature due to the polarity of the molecule, the tar does not condense in one-pot due to the non-polarity of the molecules even when the temperature is much lower than 0 °C. It starts condensing from 300 °C zone to the end use zone.

Turbines are not very sensitive to tar because it can accept hot gas for combustion and since the temperature of the hot gas is higher than the dew point of tar it can stay as vapor form. However, at temperature above 400 °C, the tars can undergo a subsequent dehydration reaction to form solid coke that not only further causes fouling and plugging but also causes abrasion of turbine blade. Therefore, the safe level of tar concentration required is even lower than that of internal combustion engine [72,73]. When the producer gas (syngas) is used for solid oxide fuel cell and secondary conversion to methanol or synthetic diesel via Fischer–Tropsch synthesis, the tar can deposit as a sticky film on the catalyst surface. As the sticky tar/coke is refractory in nature under the reaction conditions, it permanently blocks the active site of the catalysts, and thus the overall reactive site of the catalyst gradually decreases and finally the entire catalysts become inactive.

2.2.3. Ammonia

Ammonia is supposed to form from the protein material as well as from some other nitrogen containing materials present in the biomass feedstock. The animal based biomass and some plant biomass as well such as alfalfa are high protein biomass and the gasification of those feedstock results in significant concentration of NH_3 in producer gas [77,78]. In addition, the pressurized gasifier

and reducing atmosphere in the gasifier generally facilitate to form more NH_3 from N_2 and H_2 [79–81]. The ammonia in the producer gas is not sensitive to internal combustion engine and turbine operation; however, it can present in the exhaust gas and can cause the violation of environmental regulation. The NH_3 can also be converted to NO_x during combustion and the emission of which is also strictly prohibited [82]. Thus the acceptable level of NH_3 in the producer gas is dictated by the local authority to meet the requirement of emission control. However, it can cause catalyst deactivation for Fischer–Tropsch synthesis and solid oxide fuel cell application [83].

2.2.4. Sulfur compounds

Biomass inherently contains lower percentages of sulfur compared to coal. Most of the woody biomass contains less than 0.1% sulfur, while it is around 0.3–0.4% in herbaceous crops residues. The refused-derived fuel (RDF) contains even more sulfur and it may go up to 1%, almost the same as coal. Under gasification conditions of those feedstock, sulfur is often converted to hydrogen sulfide and sulfur dioxide. Since biomass contains low sulfur, the internal combustion engine and turbine are not sensitive to sulfur compounds of biomass derived producer gas. The concentration level of sulfur compounds in the biomass derived producer gas does not need additional cleanup for engine or turbine operation. However, sulfur compounds are highly sensitive to the catalysts [84–86]. They have high affinity to adsorb permanently on the active site of the catalyst. This means that the active site acquired by the sulfur compound is no longer available for the catalyst cycle, and thus after a while the catalyst becomes dead. Therefore, the process that involves catalysts such as Fischer–Tropsch synthesis and even fuel cell cannot accept the sulfur compounds containing producer gas, and thus for those applications, thorough removal of sulfur compounds is necessary.

2.2.5. Hydrogen chloride

Biomass also contains a low concentration of halides, especially chloride. The halide gases primarily convert to acids like HCl. A few reports present precise quantities of HCl in producer gas. Depending on the biomass feedstock, the producer gas contains several ppm (90 ppm) of HCl as reported in the literature [74]. In some other reports, it was found that the HCl concentration is even higher (200 ppm) in the producer gas produced from wood gasification in autothermal fluidized bed gasifier [87]. The HCl at such a level of concentration is likely to corrode the downstream equipment, and thus it is necessary to remove from the producer gas before being fed. As can be seen in Scheme 1, many researchers did not emphasize on removing hydrochloric acid [88–94]; however, some researchers put their effort to remove hydrochloric acid in order to fulfill the stringent requirement of downstream applications of product gas [95–99].

2.2.6. Alkali metals

Trace elements especially K and Na inherently present in biomass and vaporize under gasification temperature. The vapors can be readily condensed at relatively lower temperature, contributing to the fouling of working surfaces in energy conversion facilities [100,101]. In the application of producer gas for internal combustion engine, the alkali metals deposit in the gas inlet nozzle, while in the turbine operation, the metals deposit on the turbine blades, causing fouling problems [102,103]. For the safe operation of combustion turbine, the manufacturer's specification of alkali metals is <50 ppb [96]. These metals can also often adhere with bed materials in the case of fluidized bed reactor and since they are soft at gasification temperature, they lead to the agglomeration of the bed materials and reduce the fluidization rate. In addition, it was observed that they deposit at the air inlet of the fluidized bed gasifier and often narrow down the diameter, causing problems for air distribution. For Fischer–Tropsch synthesis catalyst, the alkali metals' tolerable limit is less than 10 ppb.

3. Gas cleaning methods in demonstration scale and their advantages and disadvantages

This section of the review mainly focuses on the cleaning of producer gas produced from the demonstration and pilot range of gasifiers. Normally, the gasification process produces raw gas containing a number of impurities regardless of the gasifier types. The raw gas cannot be used in any downstream applications, even not in the open burner due to emission regulation. Therefore, it is compulsory to clean up the producer gas before being fed into the downstream application. However, the extent of cleaning depends on the individual application as described in the earlier sections. Generally, cleaning procedures can be categorized as cold gas cleaning [15,104,105] and hot gas cleaning [74,106–109]. Until recently different pilot-scale gasification for different applications used varieties of gas cleaning methods, which lie in either of the categories. It was observed that most of the project used the combination of cleaning methods as described in the subsequent sections. However, the use of cyclone to separate comparatively the course particles is common for all types of cleaning methods. Generally, the cold gas cleaning methods can be divided into two sections: namely dry gas cleaning [110,111] and wet gas cleaning [112,113] as mentioned in Table 1. Most of the applications employ both of the techniques to reach the required specification of the producer gas for different applications. As Table 1 shows, the equipment involved in the dry gas cleaning includes cyclone, rotating particle separators (RPS), electrostatic precipitators (ESP), bag filters, baffle filters, ceramic filters, fabric/tube filters, sand bed filters, adsorbers and so on. On the other hand, the equipment normally used in wet gas cleaning includes spray towers, packed column scrubber (wash tower), impingement scrubbers, venture scrubbers, wet electrostatic precipitators, wet cyclones and so on.

Table 1
Classification of gas cleaning methods.

General classification of gas cleaning	Cold gas cleaning [15,106,107]		Hot gas cleaning [74,108–111]	
Specific classification of gas cleaning	Dry cleaning [112,113]	Wet cleaning [114,115]	Thermal treatment [32,116–119]	Catalytic treatment [54,58,62,64]
Type of equipment involved	Cyclone, rotating particle separators (RPS), electrostatic precipitators (ESP), bag filters, baffle filters, ceramic filters, fabric/tube filters, sand bed filters, absorbers, etc.	Spray towers, packed column scrubber (wash tower), impingement scrubbers, venture scrubbers, wet electrostatic precipitators, wet cyclones, etc.	High-temperature devices such as ceramic filter/candle filter	Primary bed in the gasifier or in the secondary reformer

In hot gas cleaning, there are two techniques normally used and many reports mentioned that hot gas cleaning is thermally more efficient compared to cold gas cleaning. This is due to the fact that in cold gas cleaning, the gas first needs to be cooled and cleaned and then further heated up to the desired temperature of the downstream application. However, in hot gas cleaning, the cleaned and hot gas can be directly used in the downstream application at the desired temperature. The hot gas cleaning consists of thermal and catalytic cleaning. In thermal cleaning, the main impurity (tar) is decomposed at relatively higher temperature (> 1000 °C) compared to the usual gasification temperature [32,114–117]. However, catalytic hot gas cleaning involves tar cracking catalysts [54,58,62,64]. The catalyst can be used either in the primary gasifier or in the secondary tar reformer. In any case, catalyst can reduce the temperature for tar cracking.

3.1. Cold gas cleaning

The gasification of biomass generally takes place in the temperature range of 800–900 °C, which is actually the exit gas temperature. However, the internal combustion engine and many other downstream applications require fed gas of ambient temperature, especially for gas density requirement. It suggests that the gasification gas cooling is an essential step; however, it may be before cleaning and after cleaning. This section will discuss the gas cleaning after cooling or the simultaneous operation of cleaning and cooling. The cleaning method employs physical and mechanical methods, which can be divided into two categories, namely dry and wet gas cleaning as mentioned in Table 1. The dry gas cleaning system does not involve water and the equipment consists of cyclone, rotating particle separators, fabric filters, ceramic filters, activated carbon based absorbers, and sand bed filters [107,108]. In contrast, wet gas cleaning system involves water and the equipment involved are wet electrostatic precipitators (ESP), wet scrubbers, and wet cyclones [112,113].

In this process, the hot gas first enters the cyclone where the course particles can be centrifugally and gravitationally settled down at the bottom and the gas along with some fine particles then enters the cooling and cleaning system. The process consists of water scrubbing and physical filtration. Water scrubbing simultaneously cools down the gas and captures the solid particles, tars and other contaminants. The contaminant gases such as NH_3, HCl, H_2S and SO_2 are highly soluble in water, and hence they can be readily dissolved in water when the gas mixture counter-currently flows through the scrubber. However, the residence time of the gas mixture in the water scrubber is a factor that sometimes reduces the efficiency of impure gas dissolution. For large-scale gasification systems, the impure gas dissolution by providing enough residence time in the scrubber is a real challenge. This is because it needs an unusually larger size of scrubber with a large amount of water supply. In addition, some of the poisonous gas can escape the scrubber, which needs further removal. Furthermore, some of the tars are non-polar and do not dissolve in water but can be condensed down as a liquid of separate phase. Therefore, it seems only water scrubbing is not efficient to completely separate tars.

Table 2 summarizes the details of cold gas cleaning including gasifier and feedstock types, equipment involved in cleaning, composition and quality of gas. A combination of physical mechanical gas purification system, consisting of a cyclone, spray tower, packed column scrubber, condenser, purification tower and two wire-mesh mist eliminators for cleaning of producer gas produced from a pilot downdraft gasifier unit of capacity 190 kW_e, was used. The gas composition and quality obtained are listed in Table 2 [118]. The disadvantage of this work is that the carbon conversion (73.91 wt%) and cold gas efficiency (53) are extremely low. The gas yield and heating value (LHV) obtained were 2 Nm^3/kg and 4.7 MJ/Nm^3, respectively. The gas composition and heating value are quite similar to the result of small-scale pilot plant gasification in a double air stage downdraft gasifier reported in [119]. The tar and dust concentration in the final clean gas, < 35 mg/Nm^3 and < 0 mg/Nm^3, were much lower than the acceptable range for an internal combustion engine. The burnable gas composition obtained was slightly better in other 54 kg/h of pellet biomass gasification system [120], which resulted in higher gas yield (2.2–2.4 Nm^3/kg) and LHV (5.6 Nm^3/min) compared to those of other studies [118,119]. The cold gas efficiency is also higher. However, the tar and particles concentrations were not mentioned. For this test, the purification tower and condenser were reengineered to maximize the removal of tar and particles.

In reference [121], a cyclone was used for course particle separation and the product gas was instantly quenched to 50 °C using water and rapemethylester (RME) solvent. In this case the gas temperature was more than 600 °C, and thus to avoid the reforming of the solvent, the water was sprayed first and then the solvent was successively used. In this cleaning system, the tar and particles were reduced from 2.0 g/Nm^3 and 2.2 g/Nm^3 to 180 mg/Nm^3 and 0.7 mg/Nm^3, respectively. However, it seems that the tar content is still higher than that of the specification of the internal combustion engine and turbine operation. In addition, the gas cleaning with expensive solvent does not seem to be economic. Instead of RME, utilization of a combination of adsorbent and sorbent in an absorber efficiently separated tar, chlorine and sulfur gases and provided a syngas that is used for dimethyl ether (DME) synthesis [122]. Better separation of tar was achieved in this process and concentration of tar reduced to < 20 mg/Nm^3.

A 75 kW two-stage down draft gasifier operated quite stably until 400 h for producing electricity using a gas engine. Heat exchanger, bag house filter and a paper police filter were used for gas cleaning [123]. The heat exchanger reduced the temperature to just above the water dew point, and then the gas was passed through the bag house filter and a paper police filter. After a long operation, the paper police filter was checked and there was no significant buildup of dust, meaning that the bag house filter efficiently removed dust. The gas was further passed through the series of heat exchanger and paper filter to cool down and remove tar. Finally, the tar level reduced to around 15 mg/Nm^3 and the gas was used to run the engine. Venture scrubber and heat exchanger were used for cooling and tar condensation along with cyclone for dust separation [124]. Finally, the gas was passed through the chiller and demister to separate condensate. The tar and particles concentration reduced to 10 mg/Nm^3 and 10 mg/Nm^3, respectively; however the heating value (4.46 MJ/Nm^3) of the gas and the overall electrical efficiency (16.1%) were quite low. A dual-fired downdraft gasifier for wood chips gasification in pilot scale (98 kg/h) used a heat exchanger, a bag house filter and a paper cartridge filter for gas cleaning [125]. The tar content in the gas at the gasifier exit was 711 mg/Nm^3, which was brought down to 35 mg/Nm^3 at the exit of the gas cleaning system, while the dust content was brought down from 1.36 g/Nm^3 at the gasifier exit to 3.2 mg/Nm^3 at the end of the gas cleaning system. The maximum cold gas efficiency was achieved to be 89.7 with 5.3 MJ/Nm^3 of gas heating value. The cold gas efficiency was quite higher than that of the reported value [116–122], while the gas heating value was almost the same (5.38 MJ/Nm^3). This indeed indicates that the energy input for both in the gasification section and gas cleaning section is quite low in this system [125].

Recently, a number of private companies, governmental agencies and Universities in Thailand have taken numerous efforts on biomass gasification based power generation in pilot and demonstration scale as shown in Table 2 under reference [126]. All of those initiatives used downdraft gasifier in different scales with individual design and commonly used cold gas cleaning methods

Table 2
Details of cold gas cleaning including gasifier and feedstock types, equipment involved in cleaning and gas composition and quality.

Gasifier type	Feedstock type with feeding rate (kg/h)	Gas cleaning equipment	Gas composition (vol%)					Tar (mg/ Nm^3)	PM (mg/ Nm^3)	LHV (MJ/ Nm^3)	Cold gas eff., (%)	Electrical eff., (%)	Ref.
			H_2	CO	CH_4	CO_2	N_2						
Fixed bed gasifier													
Fixed bed downdraft	Wood chip, 250	Cyclone, spray tower, packed column scrubber, condenser, a purification tower, two wire mesh mist eliminators	16.1	16.6	2.3	13.8	51.2			4.7	53.0	16	[118]
Double air stage downdraft gasifier	Eucalyptus wood, 10–12	Cyclone, heat exchangers, and a bag house filter	16.8	19.0	0.9	13.6	50.6	<35	<10	4.6	67.0		[119]
Downdraft gasifier	Sawdust and sunflower seed pellet, 54	Cyclone, venture scrubber, chiller condenser, two saw dust filter and a bag filter	17.2	21.2	2.5	12.2	67.7			5.6	67.7		[120]
Fixed-bed twin fired	Wood chip, 25	A cyclone and a RME (rapemethylester)/H_2O quench system followed by a wet electrostatic precipitator (ESP)	18.3	20.4	2.5	14.7	45.5	180–240	0.7	5.8	63.5		[121]
Two stage downdraft	Corncob, 45–50	Heat exchanger, bag filter, limestone+activated carbon+desulfurization sorbent packed in a absorber	25–38	25–38	<2	16–25	8.0–10.0	20	20				[122]
Two stage downdraft	Wood chip	Heat exchanger, bag house filter, paper cartridge filter, demister	32.5	15.0	2.1	19.5	30.0	15		6.0		25	[123]
Downdraft gasifier	Olive kernel, 100–110	Cyclone, venture scrubber, heat exchanger, chiller, mist eliminator, fine filter	24.1	10.7	4.2	4.6	75.0	10	10	4.46	75.0	16	[124]
Dual fired downdraft gasifier	Wood, 98	Heat exchanger, bag house filter and paper cartridge filter	21.3	20.5	1.1	10.7	89.7	35	3	5.3	89.7	21	[125]
Downdraft gasifier	Rice husk, 85	Cyclone, wet scrubber, bag filter	6.9	17.2	4.1	19.4				5.6			[126]
Downdraft	Wood chips, 50	Physical filter	9.34	29.4	0.2	9.71	73.0			5.1	73.0	10	[126]
Downdraft gasifier	Wood chips and rice husk, 80 and 120	Cyclone, water scrubber, chiller scrubber								5.52		18	[126]
Fixed bed down draft gasifier	Wood chip	Cold gas cleaning								4.6	70.0–75.0		[126]
Fluidized bed gasifier													
Dual fluidized bed steam gasification	Poplar chips, 57–92	Pilot gas cleaning using CaO absorber, cyclone and cold gas filter	33.1	25.1	10.4	19.3	70.0		2.1	12.7	70.0		[127]
Dual fluidized bed		Heat exchanger, filter and scrubber	50.6	16.5	12.9	12.3	10.0	1000					[128]
Bubbling fluidized bed	Pine, maple-oak wood, seed corn, 86–170	Cyclone, bag house filter, iso-propyl alcohol impinger	16–17	19–21	6–7	19–20				8.26			[129]
Fluidized bed	Sewage sludge, 570	Cyclone, gas cooler, granular bed filter, Ceramic filter, water absorber, packed column for NH_3 and H_2S	13.8	13.3	4.2	13	54.7			4.7	70		[130]

consisting of cyclone, wet scrubber and bag filter. Ten gasification plants are thoroughly investigated in terms of gas yield, composition, and cleaning. Most of them used internal combustion engine for electricity generation. However, they have faced numerous problems including controlling of gas yield and compositions due to the lack of a good design of feeding system and tar removal to an acceptable range. The studies suggested that continuous improvement by research is necessary to achieve the goal of biomass based power generation.

Different types of both fixed bed and fluidized bed pilot gasifiers are available at ENEA Research Centre of Trisaia, Italy, for operating gas turbine and molten carbonate fuel cell (MCFC) [127]. A 57–92 kg/h poplar wood chips feeding dual fluidized bed steam gasifier was tested to produce clean producer gas for MCFC with different steam to fuel ratios. The cleaning of producer gas consisted of both hot and cold gas cleaning methods. The CaO absorber, cyclone and ceramic filters for hot gas cleaning and a conventional filter for cold gas cleaning were used for producing MCFC grade producer gas [8]. An industrial scale (1700 kg/h, 8 MW) steam gasification of biomass in a dual fluidized bed gasifier provided very high concentration of H_2, along with CO and CH_4 [128]. This system used active bed materials (CaO), which provided the catalytic effect to convert tar to burnable gas. The tar level was reduced from 2 to 5 g/Nm^3 [11] of usual fluidized bed reactor to 1 g/Nm^3 when CaO active bed material was used. The usual cold gas filtration was used to reduce the final tar content. Although the final tar content in the producer gas is not mentioned in the paper it was reduced to gas engine acceptable ranges. Since this system used steam, the gas yield of 1.29 Nm^3/kg is lower compared to air blown gasifier; however, the heating value of the gas of the steam gasification (12.7 MJ/Nm^3) is much higher than that of air gasification due to the high concentration of the burnable gas. A bubbling fluidized bed gasifier of feeding rate 86–170 kg/h of different biomasses (pinewood, maple oak and seed corn) using oxygen enriched air and steam produced gas with different heating values depending on the biomass type and oxygen concentration in the supplied air [129]. It was observed that the burnable gas composition as well as the heating value of the producer gas increased with increasing oxygen concentration. The maximum heating value of 8.26 MJ/Nm^3 was obtained for pinewood when 40 vol% oxygen was used; however, with the same concentration of oxygen, the maximum heating value

achieved was 5.49 MJ/Nm^3 for seed corn. The average tar content was in the range of 1.7–2.0 g/Nm^3 at the exit of the gasifier. After cleaning, it was reduced to below the engine's acceptable range.

Large-scale demonstration and pilot gasification of sewage sludge were conducted at Balingen and Mannheim, Germany, to produce clean producer gas [130]. At Balingen, 223 kg/h throughput fluidized bed gasifier was commissioned in 2002, which was scaled up to 570 kg/h. Based on the experience on Balingen plant, a large-scale plant (1140 kg/h) was built in 2010 at Mannheim. Air and steam mixture was used as the gasifying agent. The gas cleaning section consists of a cyclone and a gas cooling system to simultaneously cool the gas by quenching and cleaned to produce quality gas. The gas was further passed through the granular bed filter at the bottom of the cooler, being formed by the fuel itself. About 98% of the particles and 90% of the tars were removed by the granular bed filter. To ensure the minimum tars and particles in the producer gas, it was further passed through a candle filter. To remove H_2S and NH_3, two packed columns were used after the candle filter. The final cleaned gas was used for engine to generate electricity. Although the gas was cleaned efficiently the burnable gas composition was too low and thus the heating value was 3.2 and 4.7 MJ/Nm^3 at Balingen and Mannheim, respectively, which may be due to the poor quality of the feedstock. In addition, a number of problems were identified at both the stages, which retarded the full-fledged commercialization of this technology.

3.2. Hot gas cleaning

Gas cleaning is one of the challenging steps to make producer gas useful for downstream application. Three types of impurities are usually present in biomass gasification gas: namely particles, tar and poisonous gases (HCl, NH_3, H_2S and SO_2). This section focuses on summarizing the removal of these impurities under hot conditions usually at gasification temperature. The gas filtration at temperatures above 260 °C is called hot gas filtration according to the draft of the VDI guideline 3677-3 [131]. Hot gas cleaning can be conducted either by removing the impurities by physical filtration under hot conditions or by converting the impurities especially tar to gas at high temperatures.

3.2.1. Hot gas filtration

Hot gas filtration often focuses on the removal of particulate matters and tars, with the goal of minimizing the impurities of producer gas. The Clean Air Act 1970 imposed a stringent regulation of environmental standards, which have urged the removal of contaminants from the producer gas that would otherwise be emitted to the environment as pollutants [132]. Recent development of biomass energy conversion technologies provided numerous opportunities to utilize producer gas for power generation and chemical synthesis. However, for each of the applications, producer gas needs to be cleaned as their individual specification. High-temperature particles separation is one of the improvements to the commercial application of producer gas [133]. Most of the particle removal technologies consist of cyclone [11,134] and ceramic filters [107,135–137]. However, it can be realized that the pore of the filter can often be blocked by the particles and can be build up as a filter cake, which may cause pressure drop, and it is impossible to maintain the stable pressure drop during the build-up of the filter cake. Improved techniques are thus being introduced to overcome the problems. One of such techniques is coupled pressure pulse (CPP) cleaning system [138], which ensures the regeneration of candles and efficiency of particle separation [107]. Although the ceramic candle filter can efficiently remove particles, it has very poor performance when removing tar and other contaminants. This is because, under hot gas filtration, tar remains in the gas phase and can easily escape the filter. Therefore, the cyclone and ceramic filter cannot be used for complete cleaning of producer gas but can be used in combination with other methods of contaminant removal.

3.2.2. Hot gas tar removal by thermal cracking

Simple filtration of tar using ceramic filter under hot gas conditions is inefficient. However, the tar is likely to be converted to lighter gas at gasification temperature. Therefore, hot gas tar removal often implies the tar conversion to lighter gas, which may be thermal cracking or catalytic cracking.

Thermal cracking can be defined as the decomposition of large organic molecules to smaller non-condensable gases at high temperature. The typical temperatures for thermal cracking range from 1000 °C to 1300 °C; however, the residence time required for effective cracking depends on the temperature employed [139,140]. Fraunhofer International Solar Energy (ISE) gasification process is a recent development of thermal cracking of biomass tar, employed 1000 °C maximum temperature at the partial combustion zone of a downdraft gasifier [30]. The average concentration of CO, H_2, CH_4, and CO_2 were 20–26 vol%, 10–14 vol%, 0.5–2.5 vol% and 10–14 vol% under steady state operation with tar concentration of less than 50 mg/Nm^3 and LHV 4.47–5.72 MJ/Nm^3. In the thermal cracking of tar, higher air–fuel ratio is normally used in order to maintain higher temperatures at the oxidation zone. Although the higher oxygen concentration effectively reduces the tar content in the product gas, it also reduces the gas heating value due to the deduction of burnable gas composition in the product gas. Table 3 lists the data of some recent development of thermal tar cracking in the pilot scale biomass gasification process [13,114–117,141–145]. Because of the requirement of comparatively higher temperature for thermal tar cracking, the downdraft gasifier, which can easily maintain high temperature, is commonly used as can be seen in Table 3. The concentration of H_2, CO and CH_4 normally varies from 10 to 20 vol%, 10 to 20 vol% and 1 to 5 vol%. Because of the variation of the burnable gas composition, the lower heating value of the product gas varies from 3 to 6 MJ/Nm^3. The maximum LHV (6 MJ/Nm^3) is reported in reference [141]. Meanwhile, the tar content in the product gas varied widely. Although the gas composition is quite enough to run the internal combustion engine, the tar content is much higher in most of the reported work than the acceptable range. A few papers reported the tar content to be below the acceptable range [30,143].

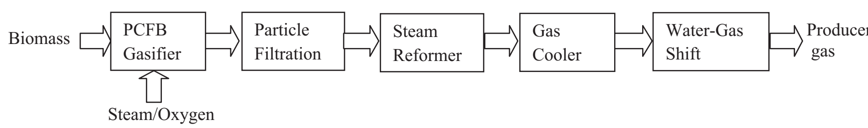
3.2.3. Hot gas tar removal by catalytic cracking

The Clean Hydrogen-rich Synthesis Gas (CHRISGAS) is the first project built on a large-scale (6 MW/9 MW) fluidized bed biomass gasification based IGCC demonstration plant in 1991–1993 at Värnamo, Sweden, and operated the plant during 1993–1999. The gas cleaning system involved the equipment for particle separation (hot gas filter), the catalytic gas upgrading reactor, i.e. steam reformer and a water–gas shift reactor [146]. The flow sheet diagram of the gas cleaning process is shown in Scheme 2. In the CHRISGAS project comprehensive studies have been carried out in the demonstration scale to produce final clean gas [147]. A number of successful test runs in the scale of 4000 kg/h biomass feeding were conducted at the IGCC plant at VVGBC during 2006 and 2007 [148]. Under this project high-temperature ceramic candle filter for particle separation [135], nickel based catalyst for tar reforming [141], and catalyst for high temperature water–gas shift reaction [149] were comprehensively tested for H_2 rich clean gas production.

Under CHRISGAS project, different types of catalysts such as magnesite, olivine and nickel based catalysts were tested for reforming of tar and increasing of hydrogen concentration in the producer gas [141,150]. Biomass type was also varied as low sulfur content such as clean woody biomass and high sulfur content

Table 3
Details of hot gas cleaning data derived from different gasifiers.

Gasifier and feedstock type	Scale (kg/h)	Temperature (°C)	Gas composition (vol%)					LHV (MJ/Nm^3)	Cold gas efficiency (%)	Ref.
			H_2	CO	CH_4	CO_2	Tar (mg/ Nm^3)			
Downdraft	Pilot, 12	1000	14.0	24.0	2.0	14.0	< 50	5.8	60-78	[30]
Downdraft	Large-scale demonstration, 5000	1000	13.0–15.0	20.0–23.0	-	10.0–11.0	–	4.2–4.3	76	[114]
Downdraft	Pilot, 18.7	900	8.7–13.2	20.8–23.6	3.6–5.2	9.3–14.5	4800	6.1	67	[141]
Updraft and Downdraft	Pilot, 30	950	10.4	15.1	0.3	12.8	450	3.2	–	[13]
Regenerative downdraft	Pilot, 5	1000	14.1–16.3	14.2–21.6	5.2–2.5	15.2–10.3	44–107	5.2–5.4	–	[142]
Down draft	Pilot	954	11.1–20.9	14.3–20.2	2.9–2.8	–	45	4.2–6.0	60-7	[143]
Downdraft	Pilot 5.4	1000–1200	11.1	18.6	2.2	11.2	3000	4.7	–	[115]
Down draft	Pilot 3–4	1050	11.11	18.56	2.0	13.12	5	3.8–4.0	63-6	[116]
Continuous Fixed bed	Pilot 4	1050	15.9	9.8	0.2	10.8	–	3.4	–	[144]
Downdraft	Pilot 2.–3.5	900–1200	17–23.3	9.9–13.5	1.5–2.8	9.9–14.5	–	4.1–5.4	63	[117]
Downdraft	Pilot 15	1100	10–12	18–22	< 1	5–20	–	4.2	–	[145]



Scheme 2. Flow sheet diagram of the CHRISGAS producer gas processing reproduced from reference [146]..

biomass such as miscanthus and straw. Miscanthus and straw also contain high chlorine, silica and potassium, which may cause agglomeration of fluidized bed materials [132,133,151]. Although olivine, magnesium iron silicate $(MgFe)_2SiO_4$ is reported to be a good catalyst for tar cracking to H_2 and CO [11,107,134,136–140] it did not provide significant impact in both reduction of tar and elevation of hydrogen in the product gas. For example, using olivine as a bed material for woody biomass gasification, the CO and H_2 concentrations were achieved to be 30.1 and 19.4 vol%, which were slightly lower compared to sand as a bed material (CO 31.7 and H_2 21.9 vol%) (Table 4) [150]. The polyaromatic hydrocarbon tar concentration in the product gas was even higher when olivine was used as the bed material, compared to sand. In contrast, some other works [30,139,140] investigated the catalytic activity of olivine for tar model compound (naphthalene) reforming in a bench scale reactor under the identical conditions of biomass gasification. They concluded that the thermal pretreatment (calcination) of olivine leads to the migration of iron species on the surface of the particle, which leads to provide the catalytic activity of naphthalene reforming. However, the kind of olivine used in the CHRISGAS project, thermal treated and untreated, provided almost similar results as the base case (using sand) in terms of gas composition and tar reduction. In contrast, magnesite ($MgCO_3$) exhibited excellent tar reforming activities, provided almost double of H_2 in the product gas by expending CO even using high impurities (S, K, Cl) containing biomass (miscanthus). The polyaromatic hydrocarbon tar (most refractory) content was reduced to around 2 g/Nm^3.

In the CHRISGAS project, commercial type nickel based catalyst ($Ni–MgAl_2O_4$) was tested for tar reforming in the secondary reforming reactor [154]. The biomasses used were saw dust, used wood and miscanthus for gasification. The reforming reaction was conducted at three sets of temperatures such as <600 °C, 750–900 °C and 900–1050 °C in order to evaluate the catalyst performance in terms of gas composition and methane and tar conversion [152]. At low temperature range, the methane conversion was negative; however, the tar conversion was positive for woody biomass. This means that methane was formed in the toluene hydrocraking as well as by the methanation reaction by consuming CO and H_2. However, in the medium- and high-temperature ranges the total syngas content significantly increased due to the fact that the endothermic methane and tar reforming reaction was thermodynamically facilitated by elevated temperature. Using magnesite in the primary bed and ($Ni–MgAl_2O_4$) in the secondary reformer, the tar reduction was achieved to the acceptable level for downstream application and total syngas content in the producer gas was as high as 70%. However, the sulfur poisoning of nickel catalyst is a big drawback, especially for straw and miscanthus. Under the CHRISGAS project, trace elements such as S, Cl, and alkali metals (K and Na) were also removed by chemical hot gas cleaning method using different types of sorbents [153]. Based on the study, the Ca and Fe-based sorbents were not suitable for effective removal of H_2S; however, Cu-based sorbent provided excellent results for H_2S removal down to 100 cm^3/m^3. For KCl separation, bauxite and keolinite were suitable sorbents to reduce KCl concentration down to 100 mm^3/m^3. However, HCl removal under hot gas condition is not suitable and thus it needs to shift to warm conditions.

Catalytic hot gas cleaning has been extensively studied by Corella and co-workers at Universities of Saragossa and Madrid (Spain). Their works were conducted in a small pilot scale (5–20 kg/h) bubbling fluidized bed reactor. The catalysts used were dolomite and commercial nickel based reforming catalysts. They used catalysts in the primary bed specially dolomite catalysts and secondary bed for nickel catalysts. Dolomite, especially calcined dolomite, in primary bed is reported to be an active catalyst for tar reforming. The in-bed use of dolomite significantly changed the product distribution at the gasifier exit because of in situ catalytic reactions of dolomite compared to the sand bed [154]. The tar content in the exit gas decreased from 12 g/Nm^3 to 2.0–3.0 g/Nm^3. In addition, the H_2 content increased from 25–28 to 43 vol%, while the CO content decreased from 45 to 27 vol%. However, the LHV was slightly deceased, mainly because of the decreasing light hydrocarbon content. The dolomite catalyst was also used in the secondary bed reformer where it performed slightly better in

terms of tar reduction (1700 mg/Nm^3) [155]. In some experiments using calcined CaO and MgO in the secondary bed, the tar content reduced to 400 mg/Nm^3 [156]. Despite the significant activity of dolomite in tar reforming to producer gas, the tar reduction never reached the required level. In addition, the catalysts were deactivated in a short time [157,158].

In order to reduce the tar content to the required level, Corella and co-worker investigated different types of nickel based steam reforming catalysts in the secondary bed reactor [159–161]. The concept of utilizing nickel based catalyst was originally from Sweden with the MINO process [162] and at PNL of Richland, WA [163–166]. They used catalysts in the primary bed where the nickel based catalysts were instantaneously deactivated due to the fouling of coke on the active site of the catalysts. Later PNL people proposed using nickel catalysts in the secondary bed downstream; however, they could not progress significantly. In the 1990s, Corella and co-worker further investigated the possibility of utilizing nickel catalyst in the secondary bed. Based on their investigation, it is concluded that nickel based catalysts can only survive when the tar concentration in the gasifier exit gas is less than 2.0 g/Nm^3 [159–161]. However, the producer gas from the conventional gasifiers often contains 2.25–42.0 g/Nm^3 [155] of tar. Therefore, they used a primary guard bed consisting of dolomite to reduce the tar content in the raw gas to their suggested concentration, 2.0 g/Nm^3. Combining the primary guard bed of dolomite and secondary reformer with nickel based catalyst the system could effectively reduce the tar content to 5–20 mg/Nm^3. Nevertheless, they commented that even though the tar content was reduced in this effective way, the system is too complex to be commercialized in a large scale.

Recent development by Asadullah and coworkers using novel metal catalyst (Rh) for total carbon conversion in biomass gasification is quite convincing [47–54]. The utilization of Rh based catalyst in the primary bed almost completely converted carbon in biomass at unusually low temperatures (650–700 °C). Different types of noble metal catalysts were prepared in their work and tested in the primary bed gasifier. Obviously, Rh/CeO_2 catalyst exhibited far better performance than the others tested [47]. However, in long-run experiments, the catalyst was deactivated due to the sintering problem. The problem was overcome by incorporating SiO_2 as a second support, which inhibited the agglomeration of the catalyst particles [50]. The catalyst was tested in different reactor systems for the gasification of different biomasses [51–54,167–169]. It was also further modified by using Ni instead of Rh [170,171]. However, the major problem related to this catalyst is the high cost associated with Rh and CeO_2. A breakthrough development by Asadullah and his coworker at Curtin University, Australia, and Universiti Teknologi Mara, Malaysia, overcomes most of the barriers related to the catalytic gas cleaning. In this work, they developed an especially designed gasifier and char-supported iron catalysts, which in combination exhibited much better performance in terms of tar reduction to less than 100 mg/Nm^3 and burnable gas composition higher than 80 vol%. This method is much better than other catalytic systems developed so far. This work has been patented [172–174] and the Curtin University together with some private companies formed a new company named Renergy [175] for further development in a larger scale with the financial support of the Australian Government.

4. Comparison of different methods of gas cleaning

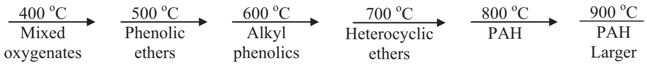
For effective downstream application of biomass gasification the gas needs to satisfy a number of essential requirements such as gas composition, tar content, particles content, gas heating value and cold gas efficiency as mentioned earlier. Since biomass based energy is still a long way from competing with fossil based energy in terms of cost effectiveness, the technologies involved in gas production and cleaning need to be as simple as possible to produce acceptable gas for individual downstream application. The efficient technologies can only provide the high quality gas with reduced cost. The subsequent sections critically compare the technologies so far developed in order to provide quality gas in terms of gas composition, tar content, particles content, gas heating values and cold gas efficiency.

4.1. Gas composition

Gas composition is one of the most important criteria of quality product gas for different applications. For power generation, the required gases are CO, H_2, CH_4 and higher hydrocarbons which are burnable. However, for Fischer–Tropsch synthesis, the CO and H_2 contents are essential. The composition of the product gas can be controlled by controlling the gasification conditions and also by post-treatment of the product gas. Higher temperature, usually above 1000 °C, favors the formation of CO and H_2. In addition, lower air to biomass ratio also facilitates the increasing formation of burnable gases including CO, H_2, CH_4 and higher hydrocarbons. However, higher temperature essentially requires expensive alloy materials for gasifier construction and complex operational systems. In addition, the lower air to biomass ratio favorably yields high concentration of unwanted tar in the product gas. Although the multistep physical filtration of either hot gas or cold gas can provide clean gas, the process is not related to change in the gas composition. Besides, because of the stringent environmental regulation and toxicity of tar, the handling and disposal of the collected tar often make it complex. In contrast, the addition of effective catalysts either in the primary bed where gasification

Table 4
Details of catalytic hot gas cleaning data derived from different gasifiers.

Gasifier and feedstock type	Scale (kg/h)	Catalyst type/non-catalytic	Gas composition (vol%)						LHV (MJ/Nm^3)	Ref.
			H_2	CO	CH_4	CO_2	N_2	Tar (mg/Nm^3)		
Fluidized bed for wood	20	Sand Primary bed	21.9	31.7	8.6	30.7	31.5	10.1	–	[152]
Fluidized bed for wood	20	Olivine $(MgFe)_2SiO_4$ Primary bed	19.4	30.1	8.9	36.0	53.1	12.8	–	[150]
Fluidized bed for wood	20	Magnesite ($MgCO_3$) Primary bed	35.9	12.7	5.5	42.6	49.0	2.2	–	[152]
Circulating fluidized bed, Wood Miscanthus	20	Ni–$MgAl_2O_4$ Secondary bed	45	17	1.8	–	–	–	–	[154]
Fluidized for Pinewood chip	Pilot 10	Calcined dolomite in bed	43	27	4.8	20	–	2000–3000	12.3	[156]
Bubbling fluidized bed for Pinewood chip	10	Secondary bed dolomite	38	36.9	7.2	33.0	–	1720	15.0	[157]
Bubbling fluidized bed for Pinewood chip	5–20	Commercial Ni catalyst	51–59	24–32	0.2–1.6	9–23		5–20	10–12	[161–163]



Scheme 3. Tar maturation scheme proposed by Elliott [176].

takes place or in the secondary tar reformer can significantly enhance the burnable gas composition even at lower temperatures. Table 4 shows the gas composition of catalytic and-non catalytic gasification/gas cleaning of biomass/producer gas. It can be seen from the data that the catalyst significantly favors the yield of burnable gas composition.

4.2. Tar content

Based on the literature, tars can be defined in many ways. Generally, it is a complex mixture of aromatic compounds with a boiling point above that of benzene and is condensable upon cooling. During biomass gasification, the organic compounds evolved below 500 °C are mostly oxygenated, which upon reforming with gasifying agents such as O_2, H_2O and CO_2 sequentially converts to gas as well as to poly-aromatic hydrocarbons. The transition of primary organic products in gasifier to final polyaromatic hydrocarbon as a function of temperature is shown in the tar maturation scheme in Scheme 3 [176]. As the temperature increased, the organic compounds become more stable, and once polyaromatic hydrocarbons form they lose their reactivity for further conversion to smaller molecules at usual gasification temperatures. The tar formation reaction is usually initiated by gas-phase free radial reaction of olefin and promoted by temperature and volatiles residence time [76]. Therefore, in order to eliminate tars from the product gas, two options can be considered. The tar can be physically removed from the product gas by filtration or can be inhibited from forming. As described in the previous section, physical filtration is not viable because of the difficulties mentioned. The temperature above 1000 °C can prohibit the progress of tar maturation reaction; however, as mentioned the equipment and process involved become very complex.

The introduction of catalyst in the primary or secondary reactor can effectively prohibit tar formation or convert the already formed tar to gaseous products. The catalyst developed for tar reforming so far can be classified into four groups such as (1) mineral, (2) nickel based, (3) noble metal catalysts and (4) iron based catalysts. As mineral catalyst, dolomite was extensively studied mostly by Corella and coworker. Olivine was also studied for it properties of tar reduction. These minerals produced H_2 rich gas with low tar content (2 g/Nm^3). However, the tar content is substantially higher compared to the required specification as mentioned in the earlier section. In contrast the nickel based catalyst reduced tar content to the desired levels. Meanwhile, the catalyst can only survive when the tar content in the inlet gas is less than 2 g/Nm^3 as reported by Corella and his group [159–161]. The noble metal catalyst can completely convert tar to gas; nevertheless, the catalyst itself is expensive [147–154]. It is noteworthy to mention that the iron based catalyst loaded on activated carbon is very active to convert tar at usual gasification temperatures, converting tar to the desired level [58–64]. In addition, the activated carbon-supported catalyst is cheaper compared to the nickel based and noble metal catalysts. Besides, the carbon based catalyst is highly resistive against deactivation.

4.3. Particle content

In the biomass gasification process, particles are inevitably generated. They may be ash particles or carbon based particles. The fluidized bed gasifier usually generates a higher concentration of particulate matter in the product gas due to the higher velocity of gas compared to the fixed and moving bed gasifier. For the downstream application of the product gas, the particulate concentration is essentially to be lower than 50 mg/Nm^3. Numerous methods are used for the separation of particles from the gas stream including hot gas cleaning and cold gas cleaning. However, the use of cyclone immediately after the gasifier is common to separate the majority of the particles. Under hot gas cleaning, ceramic candle filters are used after cyclone, while for cold gas cleaning different types of filters are used after cooling down the product gas. Water scrubber is also used for both cooling and particle separation. Interestingly, the char based secondary catalytic bed for tar reforming can also efficiently trap the particles.

4.4. Gas heating value

In the product gas H_2, CO, CH_4 and traces of C_2 and C_3 hydrocarbons are burnable gases, while CO_2 and N_2 are essentially present as non-burnable gas. The heating value of the product gas significantly varies depending on the composition of burnable gases, which also depend on the type of gasifier and gasifying agents. Actually, three important issues including tar, gas composition and heating value are related to gasifying agents in order to produce quality gases to satisfy the requirement of specific application and environmental regulations. Tar is the major factor, which actually controls the gas composition and heating value. As specified, to reduce the tar concentration in product gas to a minimum requirement, the conventional gasification system needs a higher equivalence ratio of air, which can be attributed to the higher non-burnable gas composition, and thus reduces the gas heating value. Although the addition of steam can enhance the H_2 concentration due to water–gas shift reaction, the overall heating value remains unchanged. Instead, it reduces the overall thermal efficiency of the gasification system.

Integration of catalytic tar reformer with gasifier can not only enhance the calorific value of the product gas but also reduce the tar and particles. The important features of the utilization of catalyst in the secondary reactor are that in the gasification section the lower ER can be used to produce raw gas with high tar content. Tar can be steam reformed effectively to gas in the catalytic reactor, so that the burnable gas composition could be enhanced and the dilution of product gas by nitrogen can be reduced. Hence, the calorific value is likely to be increased. However, the utilization of conventional catalysts so far developed for tar reforming is limited due to deactivation problems. For example, dolomite and nickel based catalysts have been reported to be inefficient for long-run applications due to the deposition of coke-like materials on the surface, which causes deactivation of the catalyst. The recently developed char-supported iron catalyst is found to be comparatively better because of two reasons: (1) it is more resistive against coke formation and (2) the active ingredient (Fe) and energy content in the char can be recovered by simply gasifying the spent catalyst. The resistivity of this catalyst can be explained by the fact that during tar reforming, some of the carbons of the supported char can also react with steam, producing CO, which upon evacuation creates new pores and embarks some hidden iron species, so as to create new active sites. Therefore, even some of the active sites are covered with coke; the formation of a new site can balance the activity, while the deposited coke can also react slowly to be cleaned up. This implies that the char-supported iron catalyst has the potentiality to be used for large-scale application.

4.5. Cold gas efficiency

Cold gas efficiency is one of the major criteria of typical producer gas quality. It can be defined as the ratio between the calorific value of the product gas and the total energy input including the calorific value of biomass, sensible enthalpies of biomass, air or oxygen, the enthalpy of water vapor in air, enthalpy of steam, etc. Therefore, to obtain a higher cold gas efficiency, the

burnable gas composition should be as high as possible, and the heat input should be as low as possible. It clearly implies that the higher input of gasifying agent causes higher heat consumption, resulting in lower cold gas efficiency. Moreover, the addition of steam as a gasifying agent takes even more heat as sensible heat of water, latent heat of vaporization and sensible heat of steam. Consequently, the addition of a gasifying agent is essentially to be limited to a minimum; however, under this condition conventional gasification systems yield excess of tar, much higher than that of engine specification. Taking into account the tar content, gas composition and cold gas efficiency, the integration of catalytic tar reformer is essentially required. Since, the catalyst can convert tar at much lower temperatures than that of thermal reforming of tar under limited concentrations of gasifying agent, the total heat input is much lower, providing higher cold gas efficiency.

5. Conclusions

Biomass gasification gas can be potentially used for internal combustion engine and fuel cell for power generation as well as can be used for Fischer–Tropsch synthesis to produce liquid hydrocarbon fuels. However, each of the applications of gasification gas requires a specific gas quality, which is a real challenge to achieve. Nevertheless, the technologies developed so far for quality gas production brought very significant achievement, in line with the commercialization of biomass energy. It needs a synergistic combination of developed efficient technologies. Overall, the following conclusions and recommendations can be made from the studies conducted in this review in order to lead forward the biomass conversion technologies for quality gas production.

1. The selection of a gasifier must be based on gas quality instead of ease of application. For example, fluidized bed gasifier is the most popular; however, it needs excessive use of gasifying agents in addition to fine particles of biomass. Both requirements reduce the burnable gas composition and cold gas efficiency.
2. The steps of the gas cleaning systems need to be as minimum as possible to reduce the thermal input.
3. Hot gas cleaning is more advantageous in terms of thermal efficiency compared to cold gas cleaning.
4. Simple hot gas filtration is not technologically viable to provide quality gas.
5. Catalytic hot gas cleaning can provide highly combustible gas composition with minimum tar and other poisonous gases.
6. The catalytic guard bed can enhance the durability of secondary tar reforming catalyst.

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